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ABSTRACT

Transport mirrors within the National Ignition Facility, a 192-beam 4-MJ fusion laser at 1053 nm, will be exposed to backscattered light from plasmas created from fusion targets and backlighters. This backscattered light covers the UV and visible spectrum from 351 – 600 nm. The transport mirror BK7 substrates will be intentionally solarized to absorb >95% of the backscattered light to prevent damage to the metallic mechanical support hardware. Solarization has minimal impact on the 351- and 1053-nm laser-induced damage threshold or the reflected wavefront of the multilayer hafnia silica coating. Radiation sources of various energies were examined for BK7 darkening efficiency within the UV and visible region with 1.1 MeV gamma rays from a Cobalt 60 source ultimately being selected. Finally, bleaching rates were measured at elevated temperatures to generate a model for predicting the lifetime at ambient conditions (20°C), before solarized BK7 substrates exceed 5% transmission in the UV and visible region. Over a 30-mm thickness, BK7 glass will bleach in 10 years to 5% transmission at 600 nm, the most transmissive wavelengths over the 351-600 nm regions.

Keywords: solarization, color center, BK7 glass, mirrors, hafnia/silica multilayers, gamma irradiation, bleaching kinetics

1. INTRODUCTION

The primary function of the transport mirror coatings on the National Ignition Facility (NIF) is to steer the 1053-nm laser radiation from the amplifiers to the frequency converters surrounding the target chamber. This must occur at high fluences (up to 22 J/cm^2) with minimal losses or wavefront distortion. A secondary function of the transport mirrors is to reject light backscattered from the target[1] due to Stimulated Brillouin Scattering (SBS) from 351-353 nm and Stimulated Raman Scattering (SRS) from roughly 400 – 600 nm. SBS and SRS must be prevented from propagating up the laser chain and damaging diagnostic instruments on NIF. This task was accomplished by selecting coating designs with both high 1053-nm laser-damage thresholds[2] and high transmission over the SBS and SRS wavelength regions. The spectral characteristics of a typical transport mirror are shown in Figure 1.

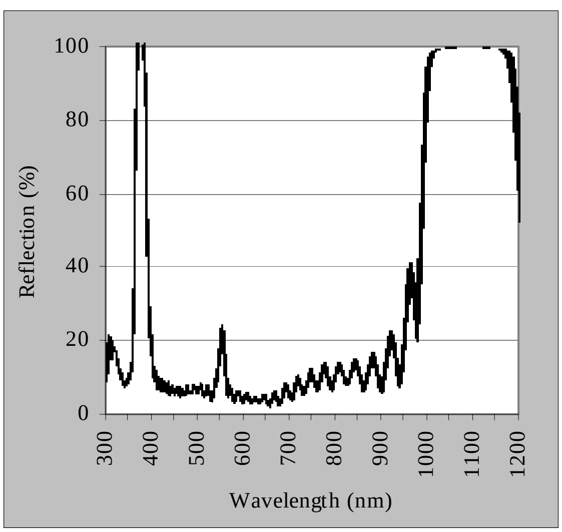

Fig. 1 Averaged spectral characteristics of 22 NIF transport mirror coating runs. The line width at each wavelength is equal to the standard deviation.

The intensity of backscattered light from the target can be as high as 2 J/cm^2. The majority of the energy will be transmitted through the mirror into the substrate and ultimately strike the metallic mounting hardware as illustrated in figure 2. The mirror mounting hardware consists of 3 expansion mandrels that are inserted in 50-mm deep holes cored into the substrate. The NIF transport mirrors are quite large with dimensions up to 525 mm × 685 mm × 80 mm. To protect the mounting hardware from laser damage, radiation-induced solarization was considered as a means of inexpensively changing the absorptive characteristics of BK7 substrates, as shown in Figure 3, to reduce the backscattered fluence to less than 100 mJ/cm^2 at the mounting lugs. This requires a 20× reduction in SBS and SRS transmission after propagating 30 mm through BK7 glass.

Radiation-induced darkening of glasses occurs when ionizing radiation generates free electrons and holes due to impurities in the glass matrix.[3] These electrons can be trapped thus creating intrinsic electron and hole color centers. Typically this is an undesirable characteristic, particularly for transmissive components such as windows and lenses. Therefore, significant research has addressed color center formation in glasses which has enabled glass manufacturers to develop solarization resistant glasses by cerium doping.[4] Since the composition of borosilicate glasses vary from vendor to vendor, the spectrum of radiation darkening has been explored for the different glass suppliers of NIF mirrors.

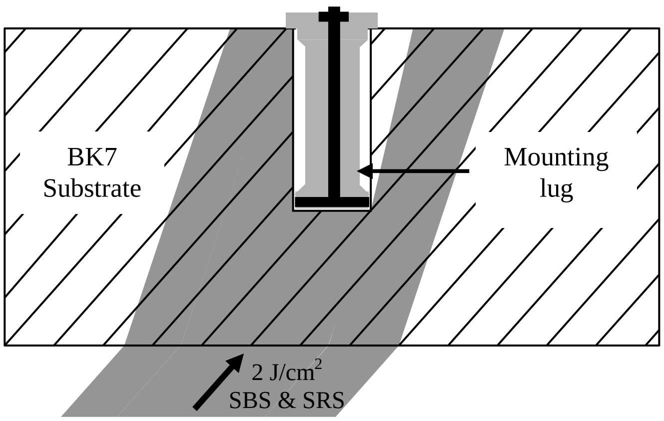

Fig. 2 Cross-sectional area of a mounting lug inside a NIF transport mirror. Backscattered light (SBS & SRS) could damage the lug if not absorbed by the solarized BK7.

Fig. 3 Solarization turns clear BK7 into absorbing glass to protect the mounting lugs from SBS & SRS backscatter. Note the three mounting holes in each mirror.

2. RADIATION SOURCE ENERGY

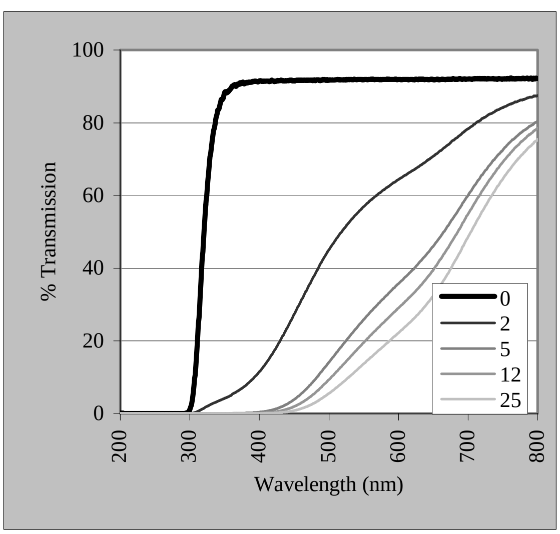

Fig.4 Spectrum of solarized BK7 exposed to 50 KeV x-rays as a function of wavelength and radiation exposure duration (in minutes).

To understand the minimum radiation required to reduce the transmission by greater than 20 times at 600 nm, uncoated BK7 glass substrates were exposed to different x-ray radiation energies ranging from 50 – 450 KeV. A BK7 sample (50-mm diameter × 10-mm thick) manufactured by Schott was irradiated at LLNL with a Rh anode x-ray tube operated at 50 KeV, and an emission current of 60 mA. The optical transmission of the BK7 sample was measured before and after irradiation using a UV-visible spectrometer (Shimadzu, model UV-1601 PC). The transmission at 351 nm (SBS) drops below 5% with less than 5 minutes of exposure as illustrated in Figure 4. As the exposure time increases, the transmission decreases. However, the magnitude of change in transmission decreases with increased exposure indicating a saturation effect. Even though the radiation darkening penetrated the entire sample thickness of 13 mm, the solarization was significantly darker at the front surface than the exit surface. Visually, the estimated thickness where the intensity of the absorption drops to 1/e is 5 mm.

The efficiency of darkening is also reduced as the wavelength increases. The highest wavelength in the SRS spectra is 600 nm. The transmission of the uncoated solarized substrate at 600 nm, including Fresnel losses, is 22%. Based on these measurements and the saturation effect, it is obvious that higher energy sources are necessary to achieve adequate darkening penetration for simultaneous blocking of both SBS and SRS backscatter.

Further tests with higher energy x-rays were conducted at LLNL using 150, 300, and 450 KeV gamma ray energy. An 80-mm thick Schott BK7 substrate that matches the NIF transport mirror thickness was used because greater darkening penetration was expected. 50-mm diameter collimated x-ray beams were used to create the solarized spots shown in Figure 5. After 3 hours, it became apparent that insufficient darkening occurs at 150 KeV. However, at both 300 and 450 KeV, there is less than 5% transmission at 600 nm. The majority of the darkening occurs within 30 mm from the irradiated surface. The 300 KeV exposed site appears darker than the 450 KeV site due to the longer duration of x-ray exposure.

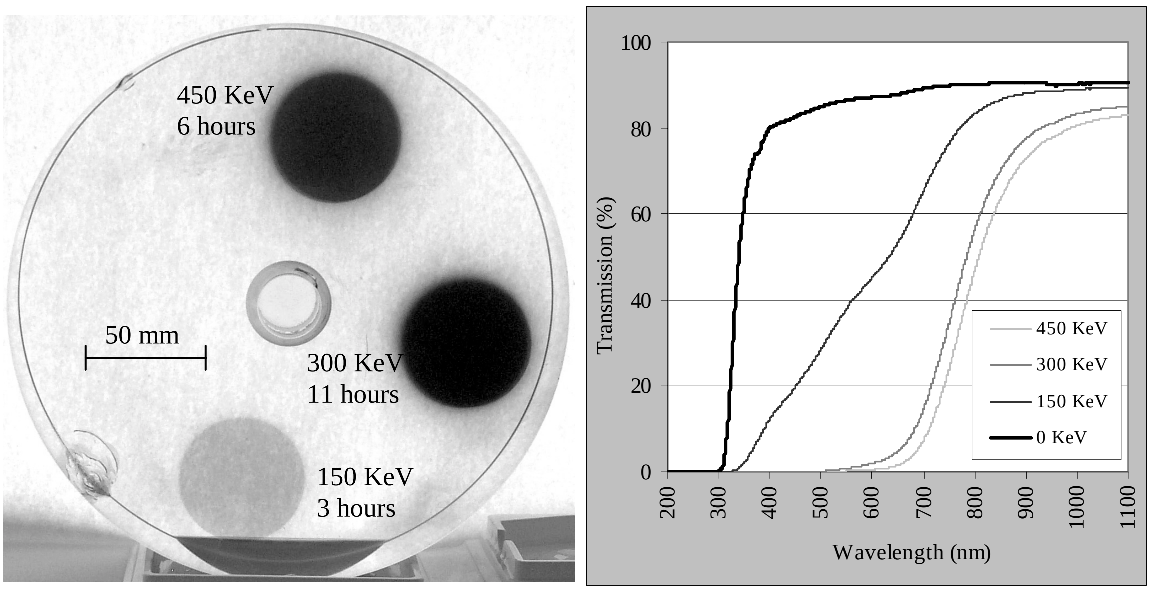

Fig. 5 Image of 80-mm thick BK7 exposed to 50-mm diameter x-ray at 150, 300, and 450 KeV (left) and corresponding spectrum (right) of the solarized regions.

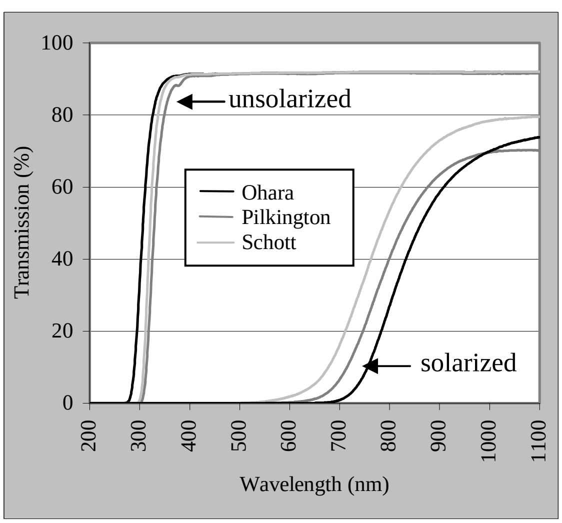

Fig. 6 Solarization spectrum of 30 mm thick borosilicate glasses from Schott (BK7), Pilkington (BSC517642), and Ohara (S-BSL7).

Commercial sterilization facilities with 1.1 MeV Cobalt 60 gamma rays exist that can easily accommodate NIF-size transport mirrors at attractive prices. The penetration of 1.1 MeV gamma rays through borosilicate glasses is 80 mm at 1/e. To understand the uniformity of darkening through an 80-mm thick BK7 substrate, 10-mm samples were stacked together to reach a total thickness of nearly 80 mm. The non-uniformity of the absorption coefficient in the UV and visible spectrum was no worse than 13% and is given by the equation:

$$\alpha = \frac{-\ell n\left(T_{solarized} / T_{unsolarized}\right)}{d}$$

NIF transport mirrors are supplied by several material vendors. Therefore, the solarization efficiency of the different materials becomes significant. Three different glasses; Schott BK7, Ohara S-BSL7, and Pilkington BSC517642 were exposed to 25 kGray of 1.1 MeV Cobalt 60 gamma radiation. Slight differences in glass composition lead to the differences in solarization efficiency as illustrated in Figure 6. These curves are based on measured optics that are only 10-mm thick. In order to meet the 5% transmission requirement at 600 nm over 30 mm thickness, a transmission of roughly 37% is needed for a 10-mm thick witness.

Ultimately the solarization needs to occur in coated optics, therefore it is important to understand when to solarize the optic during the mirror manufacturing process. Solarizing before deposition will change the thermal absorption characteristics of the substrate thus potentially impacting both coating stress and absorption. Solarizing after coating could also impact coating stress and laser damage threshold. What ultimately determines the appropriate time to solarize the optic in the manufacturing process is the rate at which the optic bleaches at elevated temperatures. At coating deposition temperatures (~ 200°C) and typical thermal cycles (24 hours), the magnitude of bleaching is quite pronounced as shown in Figure 7. A more in-depth treatment of this phenomenon is covered in section 5 where thermal annealing is used as an accelerated test to predict bleaching at ambient conditions.

Fig. 7 Impact of 1.1 MeV gamma rays on BK7. Left image is an unexposed optic. Central image is an optic exposed to 25kGray. Right image is the same optic, but bleached during exposure to elevated temperatures (200°C) typical of e-beam deposition.

3. LASER DAMAGE THRESHOLD

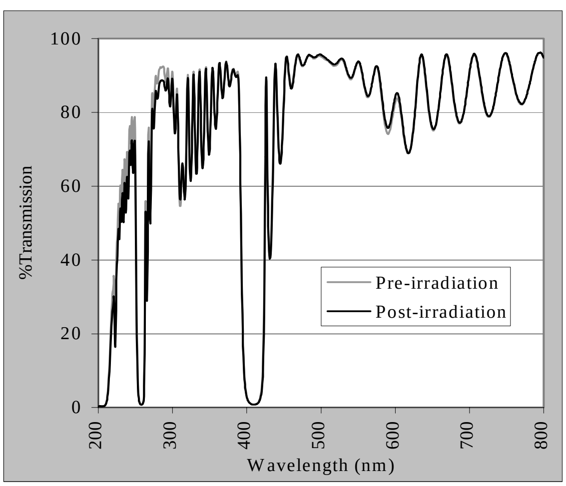

Fig. 8 Spectrum of a transport mirror coating on fused silica exposed to 2.8 Mrad of 50KeV x-rays.

Although the impact of solarization on interface absorption has been studied[5], the impact of solarization on the damage threshold of dielectric mirror coatings was unknown. A fused silica sample (50-mm diameter x 10-mm thick) manufactured by Corning (7980) was coated with a 45 degree hafnia silica mulitilayer transport mirror coating and irradiated by x-rays. Fused silica does not easily solarize providing us the opportunity to explore the effects of irradiation on the coating alone.

The fused silica optic was irradiated from the coated side for a duration of 9 minutes. All irradiation was conducted with a Rh anode x-ray tube operated at 50 KeV, and an emission current of 60 mA. Under these conditions the dose is roughly 520 rad/sec, so the coated side was exposed to a dose of about 2.8 Mrad. Figure 8 illustrates the small impact of x-ray irradiation on the UV transmission of the optical properties of the coated fused silica optic as measured before and after irradiation. The visible and IR regions show negligible changes. Although this result is promising, the critical parameter is the 1- and 3-ω damage threshold which must not be degraded given the high fluence requirements of the NIF transport mirrors.

Damage tests were also conducted on the x-ray exposed coated fused silica optic in an attempt to differentiate between irradiation-induced substrate absorption effects and potential modification of coating defects. The damage test was compared to a control sample coated on BK7 in the same transport mirror run. A raster-scan damage test was conducted at a wavelength of 1064 nm and pulse length of 10 ns. The damage threshold is defined as the fluence at which 50 μm or larger size damage is first detected. The irradiated and control sample were both examined after each scan for damage to determine the damage threshold. The fluences used in the tests were 9.8, 13.8, 18, 22.1, and 25.9 J/cm^2 scaled from 10 ns to 3 ns using τ=0.35 for the x-ray exposed sample and 18, 20, and 24.7 J/cm^2 for the control sample. The reported damage thresholds are 22.4 J/cm^2 and 24 J/cm^2 for the control and x-ray exposed samples respectively. The difference of 10% is considered insignificant compared to a damage testing measurement error of 15%.

A full-aperture mirror was also damage tested before and after solarization since small-aperture damage tests are not always representative of large-aperture damage thresholds. The optic was raster scanned over 25% of the area as described elsewhere[6] at fluences of 10, 14, 18, 20, 22, and 25 J/cm^2. The damage threshold is defined as the fluence at which 300 μm or larger size damage is first detected. Damage was observed at 25 J/cm^2, which is typical of NIF transport mirrors.[2] The optic was then solarized and damage tested over another 25% area that was previously untested. Again the damage threshold was 25 J/cm^2, however, a high density, 1 per cm^2, of plasma scalds[7] were observed compared to less than 0.003 scalds per cm^2 in the unsolarized test. Although plasma scalds have been determined to be a benign damage morphology[8], a subsequent experiment was designed to determine if plasma scalding could be reduced.

A second mirror was coated to explore whether the plasma scalding density could be reduced to unsolarized densities. After coating, the mirror was processed like all NIF transport mirrors. It was laser conditioned with 3 fluence steps; 10, 14, and 18 J/cm^2. Previous experiments have validated that laser conditioning improves the damage threshold of electron beam coatings by 2 times.[6] The mirror was then solarized and rescanned at 18 J/cm^2. No significant change was observed in the plasma scald density after solarization and raster scanning.

Damage tests were also conducted on a 50-mm diameter coating witness at 3ω. Solarized substrates and coatings have little absorption at 1ω, therefore it is reasonable to expect little differences in the laser damage threshold at 1ω. The impact on 3ω damage threshold is less certain due to the high substrate absorption and small transmission differences in a coating after solarization. Typically 3ω damage is much more intrinsic in nature enabling standard S:1 and R:1 testing. Specifically, either the entire irradiated area becomes damaged (high density of pinpoint scatter sites) or no damage occurs. This is very different than the low-density damage that occurs at 1ω leading to the significant difference in damage threshold with area tested. The 50-mm diameter coating witness was damage tested using the R:1 method. An area of 1 cm^2 was tested before and after solarization with the results illustrated in figure 9. The damage threshold is defined as the fluence at which 10-μm or larger size damage is first detected. The NIF requirement is a 3ω damage threshold of 2 J/cm^2 compared to the R:1 measured values of 7.6 and 6.4 J/cm^2 before and after solarization respectively. Finally, the unconditioned S:1 damage threshold was measured at 3ω over a 1 cm^2 area. Damage occurred at 3.0 J/cm^2 and 2.5 J/cm^2 before and after solarization, respectively. A summary of these results are listed in table 1.

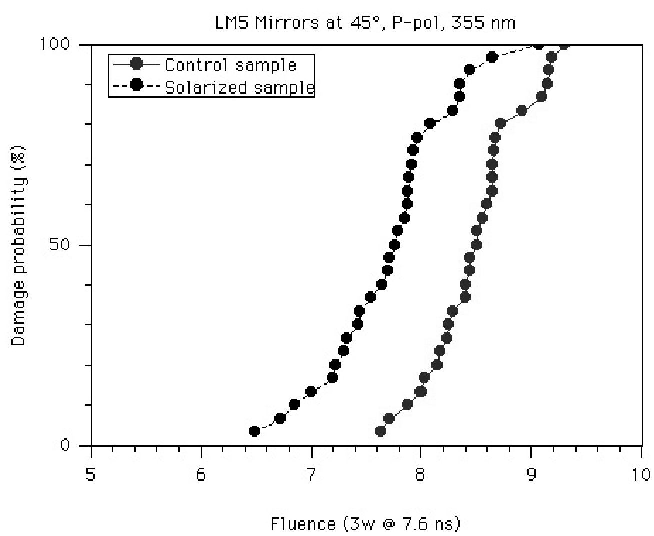

Fig. 9 Damage threshold probability curve at 351 nm of transport mirror coating before and after solarization.

Table 1 Summary of testing results suggest solarization has a negligible impact on laser damage threshold

Wavelength (in nm)	Test type	Test area (in cm^2)	Damage size criteria (in μm)	Solarized	Damage threshold (in J/cm^2 at 3 ns)
1053	Raster scan	1	50	No	24.7
				Yes	22.4
		582	300	No	25
				Yes	25
351	S:1	1	10	No	2.5
				Yes	3
	R:1			No	7.6
				Yes	6.4

4. REFLECTED WAVEFRONT

A high resolution (0.43-mm pixel size) 24" WYKO phase-measuring interferometer with a wavelength of 668 nm was used to measure the reflected wavefront of a coated NIF transport mirror before and after solarization. The reflected wavefront is directly proportional to the surface figure and coating stress. The mirror was measured at use angle, but outside of the reflection band of the coating which can lead to phase-induced errors due to minor coating thickness variations (<1% nonuniformity) across the coated optic. Therefore, only a comparison of the difference in the reflected wavefront of the mirror before and after solarization is valid. The stress of electron-beam deposited coatings is sensitive to humidity [9] so the relative humidity was at ambient conditions where the coating stress is least sensitive. The relative humidity differed by less than 5% between measurements. The P-V and RMS gradient, as illustrated in Figure 10, changed by 0.055 waves and by 0.001 waves/cm respectively at the use wavelength of NIF (1053 nm). These changes are within experimental error thus are considered insignificant.

The reflected wavefront of the mirror can also be impacted by the amount of heating that occurs when the SBS and SRS light is absorbed within the substrate. To better understand this issue, a finite element model was constructed using TOPAZ to calculate the thermal transients. These data were then used as the input of an identical finite element model in NIKE, a thermal code used to calculate the distortion.

The desired thermal recovery rate for NIF is not to exceed 4 hours. Currently the amplifier slabs limit the thermal recovery time on NIF. All other components are required to have thermal recovery times less than the amplifier slabs. The thermal calculations were done assuming 2 J/cm^2 incident energy. The mirror radiates and convects into Argon gas at ambient temperature (20°C). Conduction occurs through the mounting lugs into the rest of the mechanical structure. At 2 J/cm^2 the anticipated P-V normal displacement is 4.5 nm or 0.009 waves reflected wavefront at 1053 nm as illustrated in Figure 11.

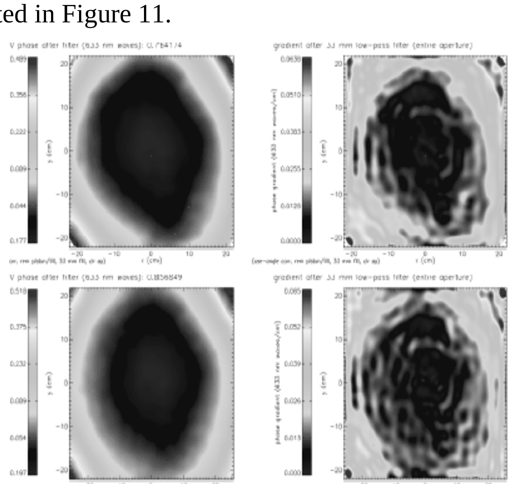

Fig. 10 Wavefront results of p-v (left) and RMS gradient (right) of before (top) and after (bottom) solarization. The p-v changed 0.055 waves and the RMS gradient changed 0.001 waves/cm.

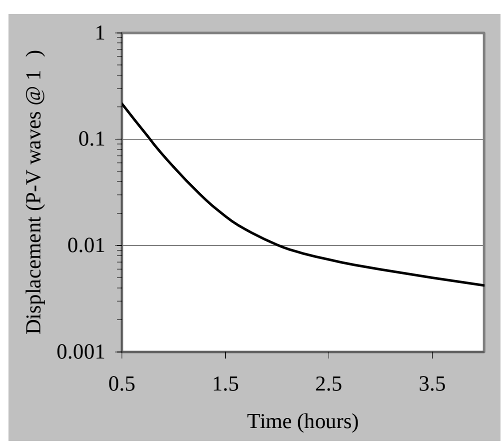

Fig. 11 The modeled surface figure distortion of a transport mirror is 4.5 nm after 4 hours. The distortion is caused by thermal effects when SBS and SRS is absorbed in the solarized mirror.

5. BLEACHING RATES

Color centers can be annihilated by releasing the trapped electrons thus holes and allowing them to recombine.[10] This process is accelerated at elevated temperature as illustrated in Figure 12. Because of this, optics can not be solarized prior to electron beam coating since such coatings are applied at elevated temperatures (~200°C).

Although the bleaching rate slows at lower temperatures, it is not negligible at ambient temperatures. To predict the lifetime of NIF transport mirrors maintaining a 600-nm transmission of less that 5% over 30-mm distance, a model consisting of a basis set of two branched second-order reactions was constructed.

5.1. Bleaching measurements

A convection oven containing two 2-kg copper (Cu) blocks was used for the BK7 bleaching trials. Prior to the bleaching tests, the Cu blocks were allowed to thermally equilibrate in the oven at the desired experimental temperature. Following equilibration, the solarized optic was quickly placed in the oven between the two Cu blocks. The temperature drop of the Cu blocks ranged from about 1°C at the low temperature bleaching trial up to about 3°C at the high temperature trial. Following the desired incubation period, the optic was removed and placed between two room temperature Cu blocks. Transmission measurements were taken on the optic following cooling.

Optical transmission measurements were taken from 1100 nm to 200 nm using a commercial UV-visible spectrometer. Sample surfaces were carefully cleaned with ethanol and lens tissue prior to taking optical measurements. Duplicate transmission measurements were taken and averaged. The absorption band at 525 nm was used to track the rate of bleaching in each BK7 optic.

5.2. Bleaching model

In order to determine an adequate predictive bleaching model, absorption was determined as a function of thermal annealing time and temperature. As illustrated in Figure 13, the bleaching rate quickly drops with time at constant temperature. Also, low temperatures were found to be dominated by one reaction rate while high temperatures had a very different reaction rate. Given the dichotomy of these results, a model with two branching second-order reactions was constructed.

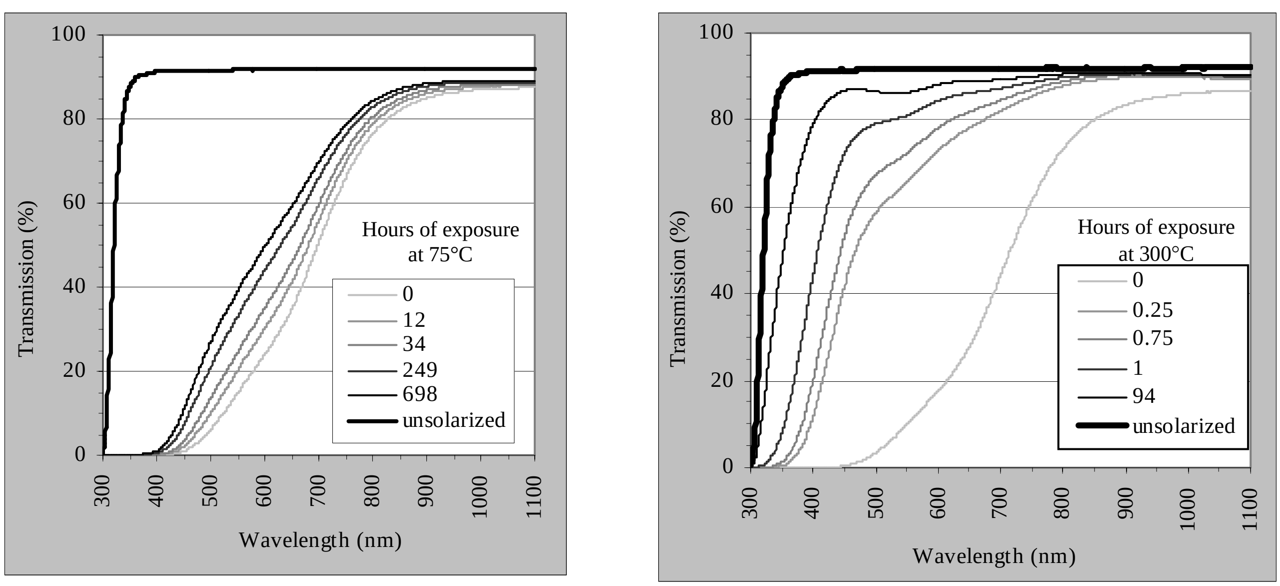

Fig. 12 Spectrums of solarized BK7 after different thermal exposure times (left image 75°C; right image 300°C).

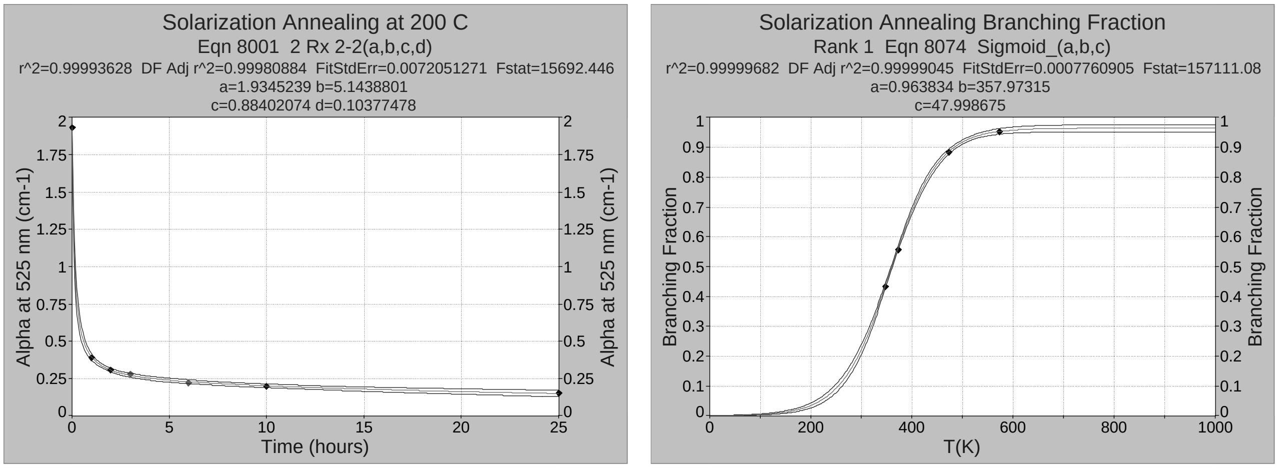

Fig. 13 Bleaching rate at constant temperature decreases with time (left graph). Branching fraction is the contribution of each of the low and high temperature reaction types considered over a range of temperatures (right graph).

The equation used to mold the thermal annealing is as follows:

$$\alpha(t) = \alpha_o \left(\frac{\beta}{1 + \beta \alpha_o K_1 t} + \frac{1 - \beta}{1 + (1 - \beta) \alpha_o K_2 t} \right)$$

where α is the wavelength and time dependent absorption coefficient (Figure 13), β is the branching fraction of the low and high temperature reactions (Figure 13), and the rate constants are $K_i = Ae^{-Ea/RT}$ (Figure 14). All of these parameters were determined by fitting the experimental data collected at each temperature to the model for each of the 3 different borosilicate glasses. Excellent agreement between theoretical predictions and experimental results was observed as illustrated in Figure 14.

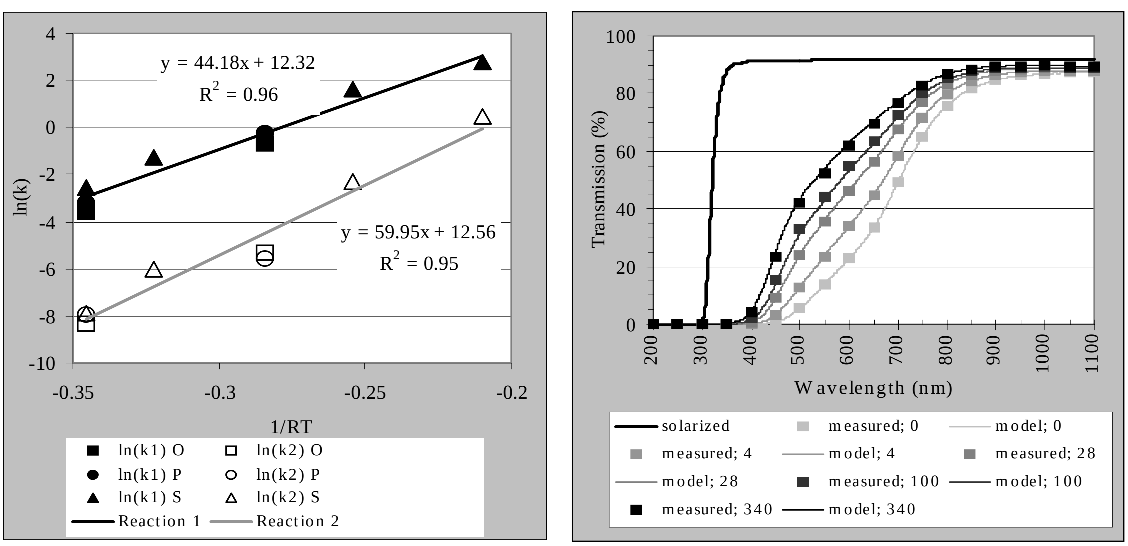

Fig. 14 Arrhenius plot of temperature dependence of rate constants of borosilicate glasses from Ohara (O), Pilkington (P), and Schott (S) for the two different reactions (left chart). Comparison of theory and experiment of bleaching rates at different time durations (in minutes) at 200°C (right image).

Using the model, bleaching rates can be extrapolated to 20°C for each of the three different glasses as illustrated in Figure 15. Low temperature bleaching studies are underway to validate the extrapolation, however, a year or two will be required between tests to get any meaningful data. According to the model, the lifetime of a NIF transport mirror before the transmission at 600 nm increases to 5% is 10 years for all three glass types. After 10 years, the mirrors will need to be removed from NIF, resolarized, and reinstalled. The expected replacement rate of transport mirrors due to laser damage during operations is approximately 10% so about half of the final transport mirrors will be replaced before resolarization is necessary.

BK7 also solarizes when exposed to 3ω (SBS) laser light. This will tend to slow the bleaching rate or reverse its effects. Given the range of different experiments on NIF resulting in a wide range of SBS fluences, it is difficult to anticipate the significance of SBS induced solarization. During transport mirror refurbishment, laser-damaged mirrors will be measured to refine the lifetime model to include the presence of SBS light.

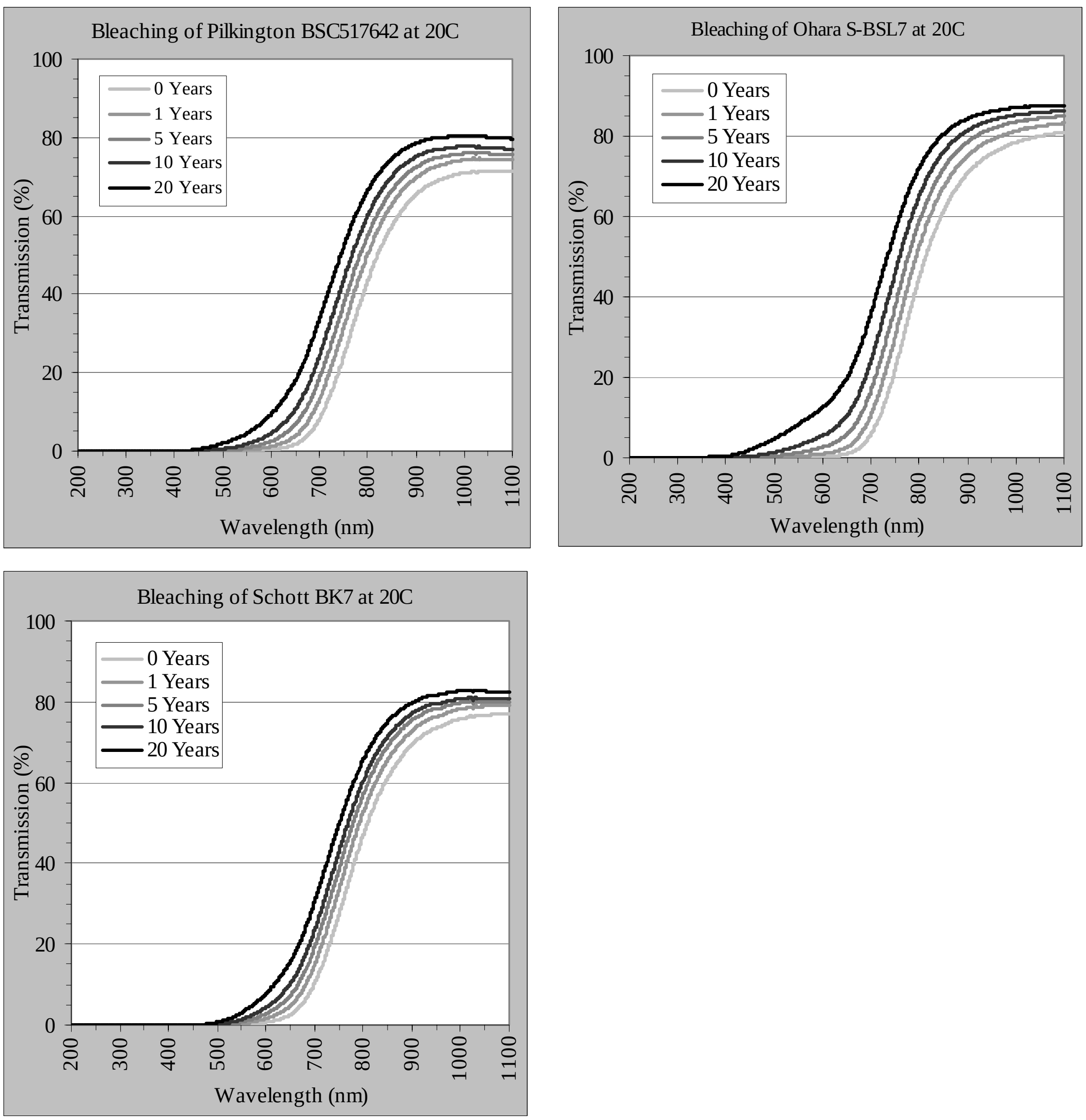

Fig. 15 Modeled bleaching rates at 20°C of 3-cm thick common borosilicate glasses from 3 different glass vendors.

6. CONCLUSIONS

Solarization is an effective low-cost method of increasing the blocking efficiency at UV and visible wavelengths. Solarization also has minimal impacts on the laser damage threshold at both 1-and 3-ω. The reflected wavefront of solarized mirrors is also not significantly altered. The solarization effect is reversible and the effect is accelerated as temperature of the substrate is increased. A model was constructed based on accelerated testing at elevated temperature to predict a 10 year lifetime for solarized mirrors before the transmission at 600 nm will increase to 5%.

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